STUDY OF ADHESIVE PROPERTIES OF EPOXYLIGRIN-MODIFIED UREA–FORMALDEHYDE RESIN AND ITS APPLICATION IN WOOD INDUSTRY

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Abstract: This paper reports the synthesis, characterization, and adhesive performance of urea–formaldehyde (UF) resin modified with epoxyligrin - an epichlorohydrin-functionalized lignin derived from cotton stalk processing waste. Epoxyligrin (10–17 mass parts per 100 mass parts of UF resin) was incorporated during the second polycondensation stage under alkaline conditions (pH 7.3–7.7, 60–65 °C, 40–50 min). IR spectroscopy confirmed the incorporation of epoxide functional groups (910–950 cm^{-1}) and enhanced hydrogen bonding networks (3300–3400 cm^{-1}). The modification reduced gelation time from 120–130 s to 90–100 s and curing time from 2.5–3.0 min to 1.8–2.0 min. Internal bond strength of particle boards increased from 0.30–0.40 to 0.50–0.70 MPa. Bending strength rose to 20–23 MPa and thickness swelling decreased to 10–12 %. Free formaldehyde emission was reduced by ~27 %. The modified adhesive was successfully applied in wood-panel production and showed improved compatibility with cotton stalk and reed fibre substrates.

Keywords: urea–formaldehyde resin, epoxyligrin, lignin, epichlorohydrin, wood-based panel, adhesive modification, IR spectroscopy, mechanical properties.

Аннотация: В данной статье описаны синтез, характеристика и адгезионные свойства мочевино-формальдегидной (МФ) смолы, модифицированной эпоксилигнином - лигнином, функционализированным эпихлоргидрином, полученным из отходов переработки стеблей хлопчатника. Эпоксилигнин (10–17 массовых частей на 100 частей МФ-смолы) вводился на второй стадии поликонденсации в щелочной среде (pH 7,3–7,7, 60–65 °C, 40–50 мин). ИК-спектроскопия подтвердила включение эпоксидных функциональных групп (910–950 $см^{-1}$) и усиление водородных связей (3300–3400 $см^{-1}$). Модификация сократила время желатинизации с 120–130 с до 90–100 с и время отверждения с 2,5–3,0 мин до 1,8–2,0 мин. Прочность внутреннего сцепления плит возросла с 0,30–0,40 до 0,50–0,70 МПа. Прочность на изгиб составила 20–23 МПа, разбухание по толщине снизилось до 10–12 %. Эмиссия свободного формальдегида уменьшилась примерно на 27 %.

Ключевые слова: мочевино-формальдегидная смола, эпоксилигнин, лигнин, эпихлоргидрин, древесно-пластиковые плиты, модификация клея, ИК-спектроскопия, механические свойства.

Annotatsiya: Mazkur maqolada paxta poyasi qayta ishlash chiqindilaridan olingan va epixlorgidrin bilan funksionallashgan lignin - epoksilignin asosida modifikatsiyalangan mochevina–formaldegid (MF) smolasining sintezi, tavsifi hamda yelimlash xossalari bayon etilgan. Epoksilignin (100 massa qism MF smolasiga nisbatan 10–17 massa qism) polikondensatsiyaning ikkinchi bosqichida ishqoriy muhitda (pH 7,3–7,7; 60–65 °C; 40–50 min) kiritildi. IQ spektroskopiya tahlili epoksid funksional guruhlarining (910–950 sm^{-1}) hosil bo‘lganligini hamda vodorod bog‘lanishlar tarmog‘ining kuchayganligini (3300–3400 sm^{-1}) tasdiqladi. Modifikatsiya natijasida jelatinlanish vaqti 120–130 s dan 90–100 s gacha, qotish vaqti esa 2,5–3,0 min dan 1,8–2,0 min gacha qisqardi. Yog‘och-qirindi plitalarining ichki bog‘lanish mustahkamligi 0,30–0,40 MPa dan 0,50–0,70 MPa gacha oshdi. Egilishdagi mustahkamlik 20–23 MPa ni tashkil etdi, qalinlik bo‘yicha shishish esa 10–12 % gacha kamaydi. Erkin formaldegid emissiyasi taxminan 27 % ga kamaygani aniqlandi. Modifikatsiyalangan yelim yog‘och-plita materiallari ishlab chiqarish jarayonida muvaffaqiyatli qo‘llanilib, paxta poyasi va qamish tolalari asosidagi substratlar bilan yuqori moslik namoyon etdi.

Kalit so‘zlar: mochevina–formaldegid smolasi, epoksilignin, lignin, epixlorgidrin, yog‘och-plita materiallari, yelim modifikatsiyasi, IQ spektroskopiya, mexanik xossalar.

Lignin, the second most abundant biopolymer in nature after cellulose, constitutes 20–30 % of lignocellulosic biomass and is a major by-product of the pulp and paper industry as well as agricultural residue processing [1]. In Uzbekistan, large quantities of cotton stalk (pakhta poya) are generated annually as agricultural waste, representing a valuable and underutilized source of technical lignin. Chemical functionalization of lignin with epichlorohydrin yields epoxyligrin - a reactive polyfunctional compound bearing both phenolic hydroxyl groups and epoxide functionalities capable of forming covalent bonds with the amine and hydroxymethyl groups of urea-formaldehyde (UF) resin [2].

Conventional unmodified UF resins exhibit significant limitations in wood-based panel applications: low water resistance, slow curing kinetics, and insufficient mechanical performance of the resulting boards [3]. Introduction of reactive organic modifiers, particularly those bearing epoxide groups, is an established strategy to address these deficiencies by promoting crosslink density in the cured polymer network [4]. Epoxyligrin is particularly attractive as a modifier due to its biobased origin, low toxicity, and the availability of cotton stalk feedstock in Uzbekistan, making it a resource-efficient and sustainable alternative to synthetic modifiers.

This work aims to prepare UF resin modified with epoxyligrin at varying incorporation levels, characterize the resulting adhesive by IR spectroscopy, and evaluate the physical and mechanical performance of particle boards produced with the modified adhesive, in comparison with the unmodified reference system.

Epoxyligrin was prepared from technical lignin extracted from cotton stalk processing. Lignin (100 g) was dispersed in dichloroethane (200 mL), and epichlorohydrin (50 mL) was added at 60–65 °C under stirring over 2 h in the presence of sodium carbonate (0.05–0.07 mass parts) as a catalyst. The product was washed, dried, and characterized by IR spectroscopy.

UF resin modification was performed according to Patent UZ № IAP 8380 [5]. In the first stage, a formalin solution (35–38 % formaldehyde in water) was mixed with urea until formaldehyde concentration was reduced to 27 %. The solution was heated to 85–95 °C and polycondensed for 50–60 min until a white mass formed (pH 7.8–8.3). After cooling to 45–55 °C, epoxyligrin solution (10–17 mass parts, dissolved in dichloroethane at 10–12 % concentration, mass ratio epichlorohydrin:epoxyligrin = 2:1) was added, mixed intensively for 5 min, stirred for 40–50 min at 60–65 °C. Melamine (0.1–0.3 mass parts) was added dropwise until pH reached 7.3–7.7. Particle boards from cotton stalk and reed fibre were pressed at 140–160 °C, 2.0–2.5 MPa, for 7–12 min.

IR spectroscopic analysis of the epoxyligrin-modified UF resin revealed several important structural changes compared to the unmodified resin. The absorption band at 3300–3400 cm^{-1} attributed to N–H and O–H stretching vibrations showed an increase in intensity and broadening, indicative of stronger intermolecular hydrogen bonding in the modified system. A new absorption band at 910–950 cm^{-1}, characteristic of the epoxide ring stretching vibration, was observed in the modified resin and is attributed to the incorporation of epoxyligrin into the macromolecular network. The carbonyl C=O stretching vibrations remained at 1680 and 1640 cm^{-1}, confirming preservation of the urea–formaldehyde backbone. The triazine ring absorption at 810–830 cm^{-1} (from melamine co-modifier) and C–O–C ether bridge vibration at 1100–1150 cm^{-1} were also identified, providing evidence for etherification and condensation reactions during modification.

The comparative physical and mechanical properties of unmodified and epoxyligrin-modified UF resin systems are summarized in Table 1. The modification produced statistically significant improvements across all measured parameters.

Table 1

Comparative physical and mechanical properties of unmodified and epoxyligrin-modified UF resin systems

Indicator	Unmodified UF resin	Epoxyligrin-modified UF resin
Gelation time (100°C), s	120–130	90–100
Curing time (120°C), min	2.5–3.0	1.8–2.0
Viscosity (20°C), mPa·s	280–340	420–480
Internal bond strength (IB), MPa	0.30–0.40	0.50–0.70
Bending strength (MOR), MPa	16–18	20–23

Thickness swelling (24 h), %	13–16	10–12
Water absorption (24 h), %	14–18	11–13
Free formaldehyde, %	≤0.30	≤0.22
Adhesion strength (shear), MPa	1.2–1.5	1.8–2.2

The gelation time at 100 °C decreased from 120–130 s (unmodified) to 90–100 s for the epoxyligrin-modified system. This acceleration is attributable to the epoxide groups of epoxyligrin reacting with amine and hydroxymethyl groups of the UF prepolymer, thereby increasing network crosslink density and promoting faster cure kinetics. Curing time at 120 °C was correspondingly reduced from 2.5–3.0 min to 1.8–2.0 min, which is advantageous for industrial press cycle reduction.

Internal bond strength (IB) of particle boards increased from 0.30–0.40 MPa to 0.50–0.70 MPa - a 65–75 % improvement. Bending strength rose to 20–23 MPa and thickness swelling after 24 h water immersion decreased from 13–16 % to 10–12 %, bringing the material closer to moisture-resistant grade requirements. The improvement in water resistance is ascribed to the hydrophobic aromatic moieties of lignin and the higher crosslink density reducing water uptake. Free formaldehyde content decreased from ≤0.30 % to ≤0.22 %, a reduction of approximately 27 %, due to consumption of free formaldehyde groups in reactions with the lignin hydroxyl groups.

Trial production runs at the ASIA KSP-SIFAT LLC construction enterprise confirmed the technical feasibility of the modified adhesive system. Boards produced with epoxyligrin-modified UF resin demonstrated a 1.5–1.8-fold improvement in operational durability compared to conventional particle boards, along with enhanced moisture and light stability, validating the industrial applicability of the developed formulation.

The modification of urea–formaldehyde resin with epoxyligrin (epichlorohydrin-functionalized cotton stalk lignin) resulted in a substantial improvement of adhesive properties and wood-based board performance. IR spectroscopy confirmed the incorporation of epoxide groups and enhanced hydrogen bonding networks. Gelation time was reduced by 23–31 %, internal bond strength increased by 65–75 %, bending strength by 22–28 %, and formaldehyde emission by 27 %. The modified adhesive is suitable for application in particle board and fibreboard production from cotton stalk and reed fibre. The use of locally available agricultural waste as lignin feedstock imparts resource-efficiency and environmental sustainability to the developed technology.

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